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Nitric acid in the stratosphere based on Odin observations from 2001 to 2007 – Part 1: A global climatology

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Abstract

The Sub-Millimetre Radiometer (SMR) on board the Odin satellite, launched in February 2001, observes thermal emissions of stratospheric nitric acid (HNO_3) originating from the Earth limb in a band centred at 544.6 GHz. Height-resolved measurements of the global distribution of nitric acid in the stratosphere between ~18–45 km (~1.5–60 hPa) were performed approximately on two observation days per week. An HNO_3 climatology based on roughly 6 years of observations from August 2001 to December 2007 was created. The study highlights the spatial and seasonal variation of nitric acid in the stratosphere, characterised by a pronounced seasonal cycle at middle and high latitudes with maxima during late fall and minima during spring, strong denitrification in the lower stratosphere of the Antarctic polar vortex during winter (the irreversible removal of NO_y by the sedimentation of cloud particles containing HNO_3), as well as high quantities of HNO_3 formed every winter at high-latitudes in the middle and upper stratosphere. A strong inter-annual variability is observed in particular at high latitudes. A comparison with a stratospheric HNO_3 climatology based on UARS/MLS measurements from the 1990s shows a good consistency and agreement of the main morphological features in the potential temperature range ~465 to ~960 K, if the different characteristics of the data sets such as altitude range and resolution are considered.

1 Introduction

Nitric acid (HNO_3) is an important chemical constituent in the stratosphere. It is one of the most abundant species of the NO_y family (NO_y=HNO_3, NO_2, NO, N_2O_5, $ClONO_2$, ...) and thus a major reservoir of active odd nitrogen (NO_x=NO+NO_2) which is responsible for the main catalytic ozone loss cycle in the middle stratosphere. The main chemical source of nitric acid is the three-body gas-phase reaction

$$OH + NO_2 + M \longrightarrow HNO_3 + M\,, \qquad \text{(R1)}$$

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major sinks are photolysis

$$HNO_3 + h\nu \longrightarrow OH + NO_2 \quad \text{(R2)}$$

and the reaction

$$HNO_3 + OH \longrightarrow H_2O + NO_3 \quad \text{(R3)}$$

(e.g. Austin et al., 1986; Brasseur et al., 1999). The molecule can additionally be formed during midwinter in the polar middle and upper stratosphere from the downward flux of mesospheric NO_x (e.g. Seppälä et al., 2004; Rinsland et al., 2005; Funke et al., 2005; Randall et al., 2006; Hauchecorne et al., 2007) through a height-dependent combination of water-ion cluster chemistry and heterogeneous conversion on sulfate aerosols involving the night-time reservoir N_2O_5 (e.g. de Zafra and Smyshlyaev, 2001; Orsolini et al., 2005; Stiller et al., 2005). Nitric acid is also known to play an important role for processes related to ozone depletion in the polar lower stratosphere. Heterogeneous chemical processes involving HNO_3 on the surfaces of polar stratospheric cloud (PSC) particles lead to the activation of chlorine (ClO_x=Cl+ClO) from its reservoir gases in the cold polar vortices during winter and to ozone loss when sunlight returns in late winter and spring. Denitrification, the irreversible removal of NO_y by sedimentation of PSC particles containing HNO_3, delays chlorine deactivation through reformation of the chlorine reservoir $ClONO_2$ during spring and may therefore lead to prolonged ozone loss (e.g. Tabazadeh et al., 2000, 2001).

Stratospheric nitric acid has been measured from the ground and from space by a variety of passive sensors operating at infrared and millimetre wavelengths. The longest data set so far is based on measurements made between 1991 and 1998 by the Microwave Limb Sounder (MLS) on the Upper Atmosphere Research Satellite (UARS) (Santee et al., 2004).

Here we present a global climatology of stratospheric nitric acid retrieved from recent observations of the Sub-Millimetre Radiometer (SMR) on the Odin satellite spanning the 6-year period from August 2001 to December 2007. In Sect. 2 we describe the characteristics of the Odin/SMR measurements and provide an overview of the observed

global distribution and the spatial and temporal variability of HNO_3 in the stratosphere. In Sect. 3, the climatological data sets obtained with Odin/SMR and UARS/MLS are compared and differences are discussed.

2 Odin HNO_3 climatology

2.1 Measurements

The Odin Sub-Millimetre Radiometer (SMR) (Frisk et al., 2003), launched in 2001, measures thermal emissions of nitric acid in a band centred at 544.6 GHz. Global fields of HNO_3 were measured between ~83° N and ~83° S on roughly one day out of three until April 2007 and on every second day since May 2007, based on 14–15 orbits per observation day and about 60 limb scans per orbit.

HNO_3 volume mixing ratios are retrieved in the stratosphere above 17–18 km (at high latitudes) with a single profile precision of about 1 ppbv (10–15 % below 30 km) and a resolution in altitude of 1.5–2 km, degrading with increasing altitude (e.g. ~3 km at 35 km) (e.g. Urban et al., 2007b). On four orbits per day, when the ground station is not available for data down-link, limitations of the spectrometer read-out data rate apply, constrained by the satellite's on-board memory, and the vertical resolution is limited to 3 km. The horizontal resolution of the limb measurements is of the order of 300 km, determined by the limb path in the tangent-layer. The satellite motion of 7 km/s leads to an additional uncertainty of the mean profile position of similar magnitude.

The systematical error derived from known instrumental and spectroscopic uncertainties has been estimated to be better than 0.7 ppbv (Urban et al., 2005c). Comparisons with measurements of other space-borne sensors such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on the Envisat satellite, the Microwave Limb Sounder (MLS) on Aura, as well as the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on SCISAT-1 however indicate a larger positive bias of the order of 2–3 ppbv (~20%) around the profile peak in the

current Chalmers version-2.0 retrievals and a small negative bias of roughly 0.5 ppbv in the range 35–45 km (Urban et al., 2006; Wang et al., 2007a,b; Santee et al., 2007a; Wolff et al., 2008). Different approaches to correct for the evident bias in the Chalmers-v2.0 HNO_3 retrievals have been explored: Wang et al. (2007a) suggested to simply correct for an average altitude offset of 1.5 km in order to improve agreement with MIPAS HNO_3 retrievals, Urban et al. (2006) published a linear fit to operational MIPAS and Odin retrievals ($VMR_{MIPAS}=0.77039\times VMR_{Odin}+0.48e\text{-}9$) and Brohede et al. (2008) used a quadratic fit to HNO_3 measurements from ACE and Odin after applying a shift of 1 km for deriving a correction term ($VMR_{ACE}=1.11\times VMR_{Odin}-0.026e9\times VMR_{Odin}^2$). In this study, we decided to shift the profiles upward by 1 km to eliminate the altitude offset and to apply the simple linear correction suggested by Urban et al. (2006). This correction leads globally to an excellent agreement with ACE-FTS measurements within 0.5 ppbv in the 19–35 km range or better 10% between 19 and 30 km, comparable with the results obtained by Brohede et al. (2008) (not shown).

Only Odin/SMR level-2 profiles with good quality (assigned quality flag: QUALITY=0) were used. The measurement response, provided in the level-2 files for each retrieval level, was required to be larger than 0.67 in order to exclude altitude ranges where a priori information, used by the "Optimal Estimation" type retrieval algorithm for stabilisation, dominates the retrieved mixing ratios (see Urban et al., 2005c, 2007b, for details).

2.2 Global distribution

The seasonal evolution of the global distribution of HNO_3 observed by Odin/SMR is shown in Fig. 1 for 6 years of observations between 2001 and 2007 and selected levels of potential temperature in the stratosphere. Individual profile measurements were linearly interpolated on potential temperature levels before being averaged in 10° wide equivalent latitude bands on each observation day. Equivalent latitude is the latitude of a given potential vorticity contour if it was centred around the pole enclosing the same area as the original contour. Equivalent latitudes thus provide a coordinate system

relative to the polar vortices, the latter being characterised by high values of potential vorticity and consequently high equivalent latitudes. We used potential vorticity data obtained from the European Centre for Medium-range Weather Forecast (ECMWF), scaled according to Lait (1994), to derive equivalent latitudes.

In the lower stratosphere at the potential temperature level of 520 K (just above ~20 km or 50 hPa), the measurements show an increase of the mixing ratios from low to high equivalent latitudes and a strong seasonal cycle at high latitudes with build-up of HNO_3 during fall and decrease during late winter and spring, as expected from gas-phase chemistry (e.g. Austin et al., 1986; Santee et al., 2004). In the Southern hemisphere, the cycle is interrupted inside the Antarctic vortex by a strong depletion of HNO_3 during June which is completed in July and can be associated with cold temperatures and the formation of polar stratospheric clouds implying removal of HNO_3 from the gas-phase. At the same time, a collar of HNO_3 rich air is observed at and around the vortex edge. Whilst relatively low values of HNO_3 inside the Antarctic vortex persist until the vortex break-up, thus indicating denitrification, this effect is not observed in the Northern hemisphere where notably low values of HNO_3 were only measured at this level on a few occasions and during relatively short periods, e.g. in December 2002, December 2004–January 2005 (see also Urban et al., 2006), and February 2007 during relatively cold Arctic winters.

In the middle stratosphere on the 1200 K level (~36–38 km), the observed global HNO_3 field is characterised by maxima occurring with considerable inter-annual variability inside the winter polar vortices mainly during July–August in the Southern hemisphere and during December–January in the Northern hemisphere, and elsewhere by low mixing ratios below the noise level of about ±0.5 ppbv in the daily averages at this level.

Further down, on the 840 K level of potential temperature corresponding to approximately 30 km, the HNO_3 fields inside the polar vortices show first minima during late fall and early winter, caused by subsidence of HNO_3 poor air from higher levels, and then maxima similar to those seen on the 1200 K level, but slightly delayed in time. In

the northern hemisphere the enhancements are not necessarily confined to the polar vortex and high latitudes. Mixing ratios outside the vortices are higher at mid-latitudes than in the tropics. At middle and low latitudes the HNO_3 field shows a time-varying asymmetry with respect to the equator which might be attributed to a modulation due to tropical oscillations such as the semi-annual (SAO) and quasi-biennal oscillations (QBO), as suggested earlier by Kumer et al. (1996) who noted a hemispheric asymmetry in measurements of the Cryogenic Limb Array Etalon Spectrometer (CLAES) onboard the UARS satellite.

2.3 High equivalent latitudes

The vertical structure and seasonal evolution of the observed nitric acid fields at high equivalent latitudes (larger than 70°) is presented in Fig. 2 for the Southern hemisphere and in Fig. 3 for the Northern hemisphere. The measurements show clearly: (1) a main layer of HNO_3 between roughly 10–15 and 50 hPa (~20–30 km or ~500–800 K) with a maximum around 25–30 hPa (23–25 km or ~600 K); (2) a strong seasonal cycle with maxima during late fall/early winter, with the southern maximum in May being slightly larger than the northern maximum in December; (3) a considerable depletion of HNO_3 in the Antarctic from June to October–November in the lower stratosphere below ~15 hPa (or 650–700 K); (4) middle and upper stratospheric HNO_3 enhancements forming above ~10 hPa (~30 km or ~800 K) typically in July–August (SH) and December–January (NH), characterised by a considerable inter-annual variability; and (5) a gradual descent of the high altitude HNO_3 enhancements during winter, joining the main layer in the periods August–October (SH) and February–March (NH). The bottom plots in Figs. 2 and 3, representing the differences from the average profile, nicely show the alternation of downward transport of HNO_3 poor air through the middle stratosphere in the beginning of the winter and of HNO_3 rich air after chemical formation: a "reversed tape-recorder effect", with downward transported air carrying the signature of middle stratospheric HNO_3 formed above 10 hPa during a short mid-winter period, thus contributing to the NO_y budget of the lower stratosphere with a delay determined

by the vertical transport rates. For a more detailed discussion of the Odin observations of enhanced HNO_3 in the middle and upper stratosphere and the connection to the variability of solar activity and meteorological conditions, the reader is referred to Orsolini et al. (2008).

3 Inter-annual variability and comparison with UARS/MLS climatology

The seasonal cycle and inter-annual variability of the Odin measurements of nitric acid in the Northern and Southern hemisphere are presented for potential temperature levels in the lower stratosphere (520 K), and in the middle stratosphere (960 K) in Figs. 4 and 5.

The Odin climatology is compared to a global climatology of nitric acid inferred from measurements of the UARS/MLS experiment between 1991 and 1998. The MLS version 6 HNO_3 profiles, retrieved from limb observations of a small spectral feature at 205 GHz, are characterised by a horizontal resolution typical for limb sounding of the order of 400 km, a vertical resolution ranging from 4.5 to 10.5 km within the pressure range 100–4.6 hPa (~16–36 km), a single-profile precision between 1 and 1.5 ppbv, and an estimated accuracy around 2–3 ppbv. Frequent 180° yaw manoeuvres of the UARS satellite, about 10 times per year, allowed to alternately cover high latitudes of both hemispheres (~80° S to ~80° N) despite the inclination of 57° of the UARS orbit (Livesey et al., 2003; Santee et al., 2004).

Figure 4 shows the seasonal cycle of HNO_3 in the lower stratosphere (520 K) for the different years of Odin observations compared to the 7-year record from MLS, represented by its $\pm 1\sigma$ standard deviation. In the tropics (equivalent latitudes lower than 20°), mixing ratios are low (<5 ppbv) with the corrected Odin measurements being slightly higher than MLS measurements by 1–2 ppbv. At mid-latitudes between ~20–60° the agreement between the two data sets is excellent. The amplitude of the seasonal cycle increases towards high latitudes with the southern high latitude maximum in May being slightly higher than the northern maximum in December. At high

equivalent latitudes (>60°), a considerable inter-annual variability is found in the Odin measurements in both hemispheres during winter and spring (SH: June–November; NH: December–March). In the Southern hemisphere, the inter-annual variability observed by Odin is more pronounced than in the MLS observations during the periods when HNO_3 mixing ratios are particularly low, both in the onset phase of denitrification during June and in the recovery phase from August to November. Moreover, Odin data show slightly larger minimum values of HNO_3 during July than MLS. The seasonal HNO_3 maximum in April–May observed by Odin is on the other hand slightly lower than the MLS maximum. In the Northern hemisphere, mid-winter values observed by Odin are systematically slightly lower by 2–3 ppbv than the range indicated by the MLS climatology. In the vortex edge regions (equivalent latitudes of 60°–70°), the agreement between the climatologies in terms of variability is very good in both hemispheres, with Odin values being slightly lower in the Northern hemisphere. Besides considering the estimated overall accuracy of the MLS and (bias-corrected) Odin climatological data sets of 2–3 ppbv and 0.5–1 ppbv, respectively, the observed differences might also partly be attributed to the better vertical resolution of the Odin/SMR measurement of 1.5–2 km compared to the coarser ~6 km for UARS/MLS at 520 K, which might lead to inclusion of nitric acid rich or poor air from lower or higher levels into the retrieved mixing ratios. This effect is most critical in the presence of strong vertical profile gradients, namely for the pronounced HNO_3 main layer at high latitudes.

At 960 K, the highest level of the MLS climatology, the Odin measurements agree generally well with the MLS climatology showing no or only a very small positive bias, except for the highest equivalent latitudes where Odin data seem to be slightly lower than MLS data during early winter. Particular enhancements in the Odin HNO_3 data at this level were frequently observed, namely during July–August 2003 and 2005 in the Southern hemisphere and during December–January in the Northern hemisphere with the winters 2001–2002 and 2003–2004 being most significant. In contrast, the MLS 7-year climatology from the 1990s does not show a significant enhancement or increase of variability during these periods on the 960 K level, which might be attributed

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to the degrading vertical resolution and sensitivity of the UARS/MLS measurements in the middle stratosphere.

The annual variability of stratospheric HNO_3 at high equivalent latitudes (70–90°) in both hemispheres is shown for various levels between 420 and 1600 K in Fig. 6.

On the lowest levels (420 and 465 K) Odin data are relatively sparse, since these levels are very close to the instruments lower measurement limit for HNO_3, with most data obtained inside the Antarctic polar vortex. The agreement between Odin and MLS is here fairly good during June/July to September with very low mixing ratios below 5 ppbv every year, compared to up to ~13 ppbv measured by MLS during May on the 465 K level. Odin mixing ratios in the lower stratosphere are larger than MLS values during October with the agreement becoming better in November due to increasing MLS mixing ratios. At northern high equivalent latitudes, low values of nitric acid are mainly found in the winters 2004–2005 and 2006–2007, whilst the available data do not indicate a significant depletion of HNO_3 in the other years.

The depletion of HNO_3 in the Antarctic vortex during winter and spring (June–November) can also be seen on the 520 and 585 K levels. Above, on the 655 K and 740 K levels, Odin measurements show a less pronounced seasonal cycle compared to MLS measurements, which is most likely caused by the limited altitude resolution of the MLS measurements. The latter show the signature of denitrification up to 740–840 K, whilst Odin observations contain this signal up to maximal 655 K. In the Northern hemisphere, mid-winter maxima between 465 and 655 K are systematically lower than the range indicated by the MLS climatology, whilst the agreement appears to be generally excellent during summer (April–October).

Finally, at the highest levels, shown here between 960 and 1600 K, particular enhancements in the Odin HNO_3 data were frequently observed during July–August in the Southern hemisphere and during December–January in the Northern hemisphere. The signal might be followed down to the 655 to 740 K levels (before it joins the main HNO_3 layer) and contributes there to the observed range of variability.

4 Summary and conclusions

Measurements of the Odin satellite provide a quasi-continuous 6-year climatological data set of nitric acid in the stratosphere from August 2001 to December 2007. Observations are still ongoing at the time of writing. We have presented an overview of the global distribution and temporal evolution of stratospheric HNO_3, based on corrected version 2.0 level-2 data.

HNO_3 increases from the tropics towards the poles, where it shows a pronounced seasonal cycle with maxima in late fall/early winter. Particular morphological features of the spatio-temporal distribution are the strong depletion from June to October–November in the lower stratosphere of the Antarctic polar vortex as well as the high nitric acid mixing ratios found in the middle and upper stratosphere during mid-winter due to downward transport and heterogeneous conversion of NO_x rich mesospheric air (de Zafra and Smyshlyaev, 2001).

A comparison with a climatology derived from UARS/MLS measurements in the 1990s (Santee et al., 2004) results in a fairly good agreement if systematic differences of the measurements are considered, in particular the considerably better altitude resolution of Odin/SMR as well as the slightly different altitude ranges of both instruments, with Odin reaching higher up and MLS lower down in the stratosphere.

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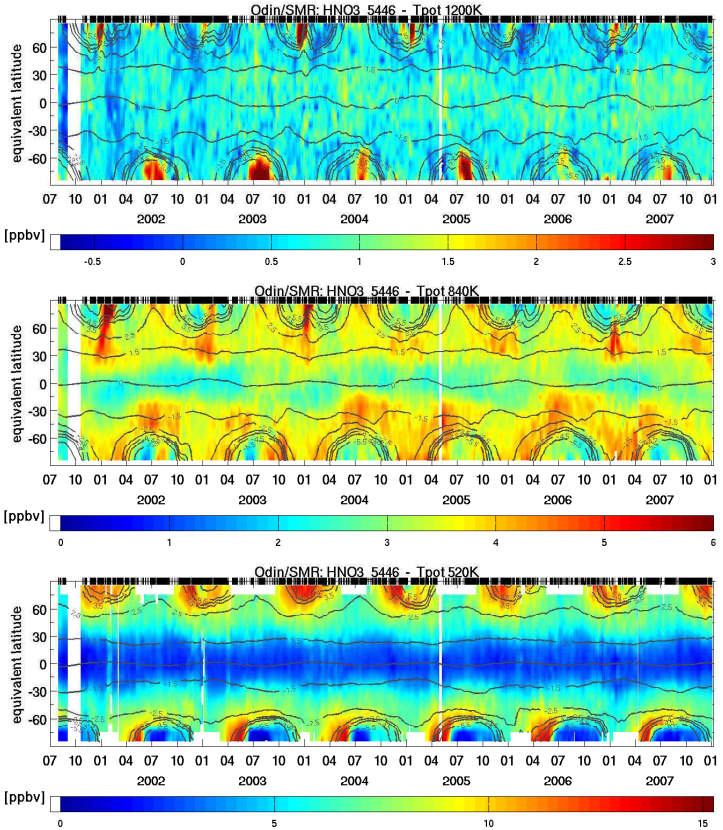

Fig. 1. Seasonal evolution of the global distribution of Odin/SMR daily zonal mean HNO_3 on the 1200 K (top), 840 K (middle) and 520 K (bottom) levels of potential temperature as a function of equivalent latitude. Grey lines indicate contours of scaled potential vorticity. Data, averaged in 10° wide equivalent latitude bands, have been slightly smoothed in time (using a 7-day moving average filter) in order to fill the gaps between individual Odin/SMR stratospheric mode observation days, indicated by crosses at the top.

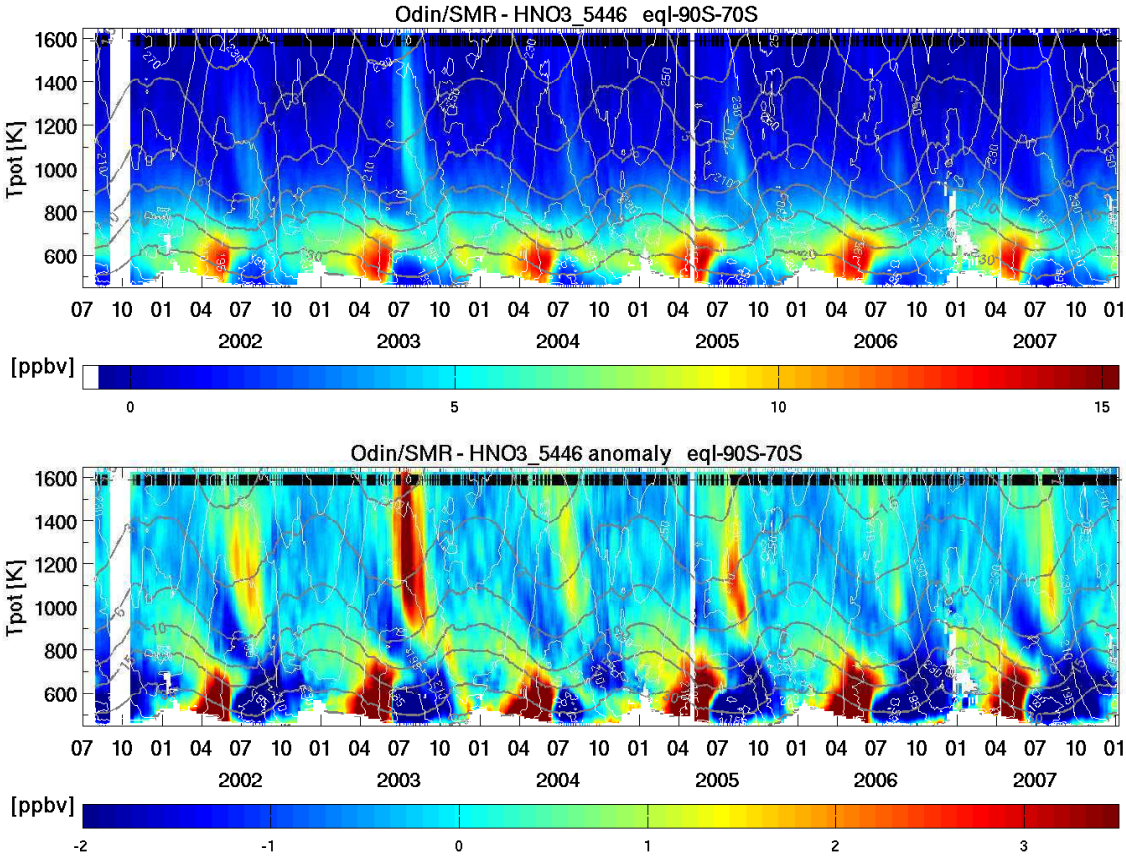

Fig. 2. Seasonal evolution of Odin/SMR daily mean HNO_3 at high equivalent latitudes south of 70° S as a function of potential temperature. Top: HNO_3 volume mixing ratio. Bottom: Absolute deviation from the mean profile. Crosses at the top of the plots indicate observation days. Pressure and temperature (from ECMWF), averaged corresponding to the Odin/SMR measurements, are superimposed using grey and white contour lines.

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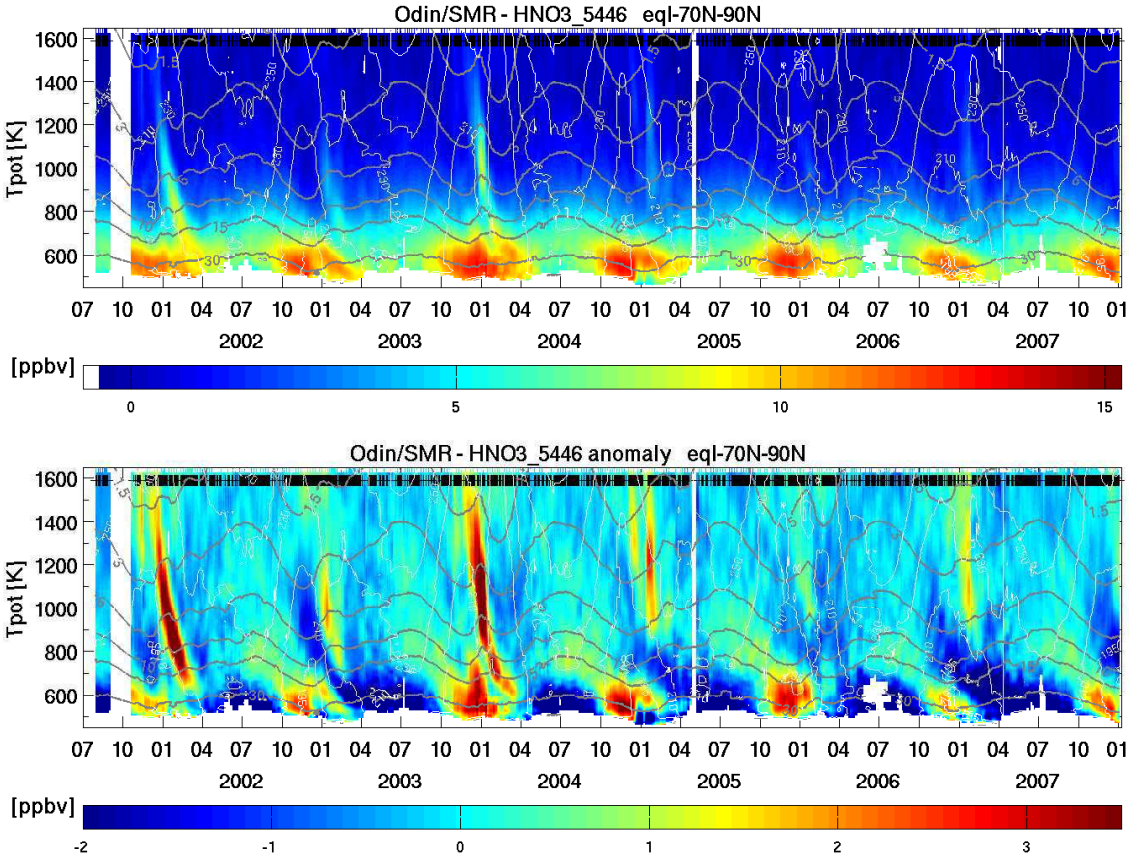

Fig. 3. Same as Fig. 2, but for Northern hemisphere high equivalent latitudes north of 70° N. Top: HNO_3 volume mixing ratio. Bottom: Absolute deviation from the mean profile.

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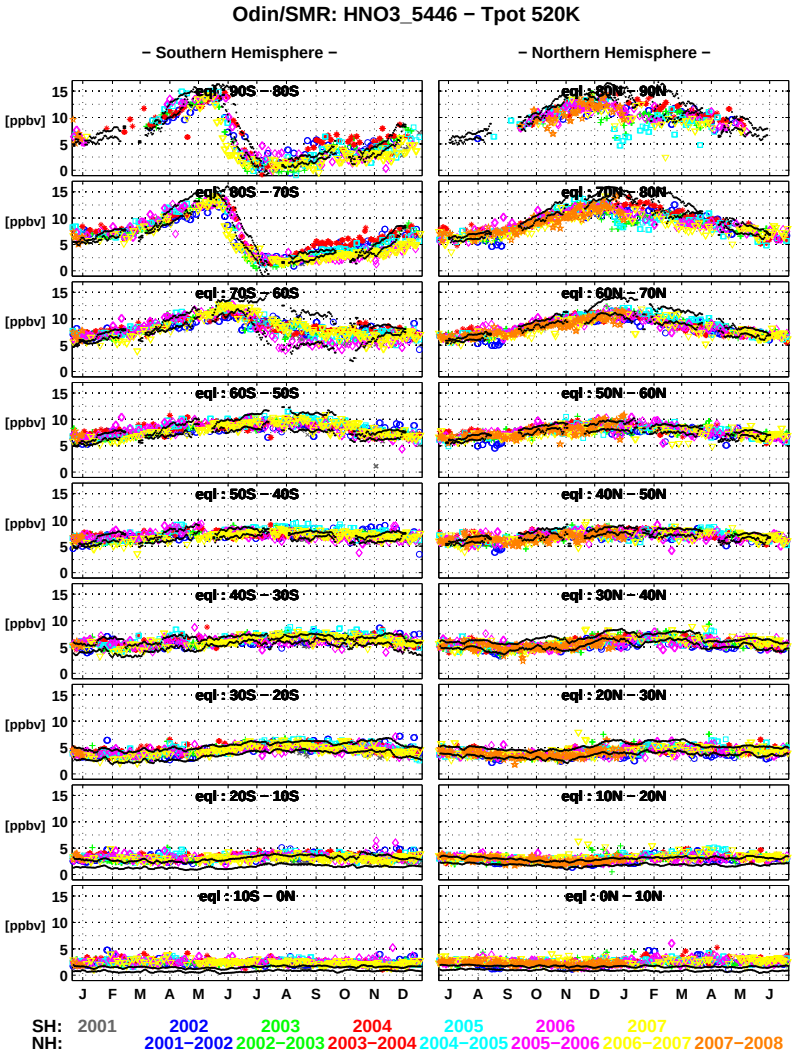

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Fig. 4. Inter-annual variability of Odin/SMR daily zonal mean HNO_3 interpolated on the 520 K level, compared to a climatology based on measurements by the MLS instrument on UARS in the 1990s. The different years of Odin measurements are colour coded (see legend). The black lines represent the $\pm 1\sigma$ standard deviation of the UARS climatology and gaps at high equivalent latitudes are due to frequent 180° yaw manoeuvres of UARS (Santee et al., 2004). Note that the x-axis range was chosen that the winter solstices are in the centre of the plots for both hemispheres.

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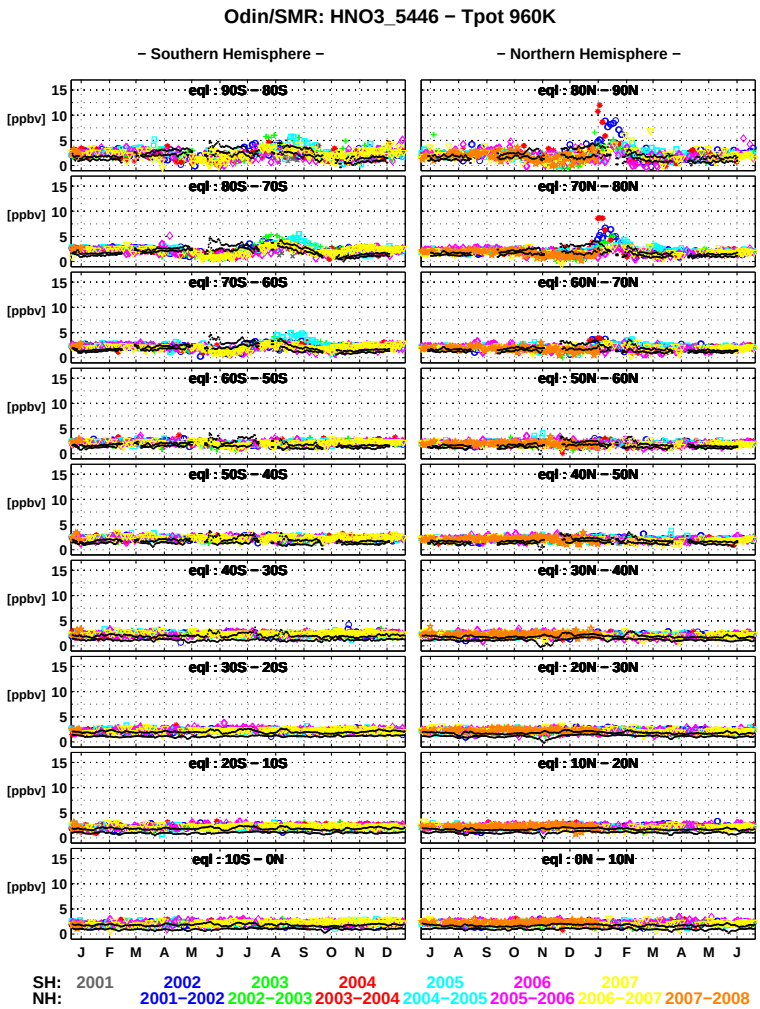

Fig. 5. Same as Fig. 4, but for the 960 K level in the middle stratosphere.

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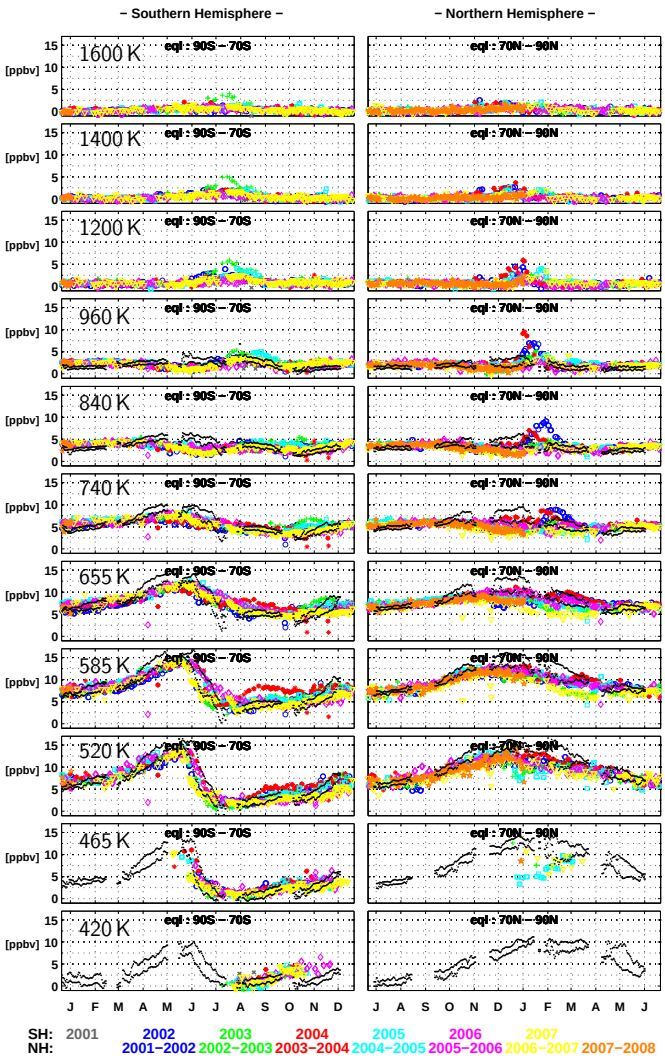

Fig. 6. As Fig. 4, but for various levels in the 420–1600 K range at high equivalent latitudes (70–90°). Whilst UARS/MLS data are available up to 960 K only, Odin/SMR covers higher levels but provides only limited information below ~500 K.

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